Elementary Excitations in Magnetically Ordered Systems with Orbital Degeneracy

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Abstract

The Holstein-Primakoff transformation is generalized to develop a quantum flavor wave theory for spin systems with orbital degeneracy. Elementary excitations of ordered ground states consist of spin, orbital , and spin-orbital waves. Spin and spin-orbital waves couple to each other due to orbital anisotropy and Hund's rule, resulting in new modes observable by inelastic neutron scattering. In the $SU(4)$ limit, flavor waves are dispersionless along one or more directions, and give rise to quantum fluctuations of reduced dimensionality.

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The Hubbard model with a Hilbert space of only one atomic orbital per site has been a popular model used to study strongly correlated electronic systems. When the correlation U is sufficiently large, the system undergoes a transition into a Mott insulator. In the large U limit, the model reduces to the familiar $S = 1/2$ Heisenberg antiferromagnetic Hamiltonian (HAH). In spite of the success and popularity of the single orbital Hubbard model, it cannot, however, explain the magnetic behavior of many Mott insulators, including ferromagnetism, magnetic ordering patterns, such as that in V_2O_3 [1–3], which would require unrealistically large long-ranged coupling if explained by HAH, and paramagnetic behavior on lattices where the HAH is known to exhibit long-range order (LRO) [4]. On the other hand, such behavior can be easily understood if one allows for orbital degeneracy and enlarges the Hilbert space on each site [5–7]. Recently, direct evidence for orbital degrees of freedom and ordering in some magnetic systems have been obtained by form factor analysis of x-ray diffraction [8].

In the case of spin systems with two-fold orbital degeneracy, the Hamiltonian is quadratic in operators S_n^m , the generators of $SU(4)$ Lie algebra. Indeed the Hamiltonian may have $SU(4)$ symmetry [7,10], rather than just the spin $SU(2)$ symmetry. In many 2D lattices such as the square or triangular lattice with nearest neighbor (n.n) coupling, the $SU(4)$ antiferromagnetic (AF) Hamiltonian has no LRO even at $T = 0$, as indicated by mean field theory, variational calculations [7], and recent Monte Carlo simulations [9]. LRO may be stabilized in higher dimensions. In real systems, the $SU(4)$ symmetry is at best approximate due to orbital anisotropy and Hund's rule. With sufficient deviation from the $SU(4)$ limit, LRO may be attained even in $2D$.

In this paper we investigate the elementary excitations of the orbital-spin systems *assuming* the ground state has broken symmetry. In effect, we are studying for orbital-spin systems the equivalence of the familiar spin waves, henceforth called flavor waves. For spin only systems, the Holstein-Primakoff transformation (HPT) maps spin operators into boson operators, and the linearized spin wave theory is equivalent to a non-interacting boson problem. For orbital-spin systems, we show that the $SU(4)$ algebra can be exactly reproduced

by a generalized HPT involving three bosons ($N-1$ bosons for $SU(N)$). A quantum flavor wave theory is then developed. We find that in the $SU(4)$ limit, even though the underlying lattice and ordering pattern is two-dimensional (2D), the flavor wave excitations can be one-dimensional (dispersionless along one direction) or localized (dispersionless in all directions). Quantum fluctuations of these excitations give rise to disordering effect of reduced dimensionality which provides further support for the lack of LRO [7,9]. The excitations in the $SU(4)$ limit can be characterized as pure spin waves (spin rotation only), pure orbital waves (orbital rotation only), and pure spin-orbital waves (simultaneous spin and orbital rotation). The anisotropy and the Hund's rule break the $SU(4)$ symmetry, and the spin and spin-orbital waves are in general mixed. We use a simplified model relevant to V_2O_3 to illustrate the mixing, and predict new modes observable in neutron scattering experiments.

With two orbital degrees of freedom, we can define orbital operator $\vec{\tau}$ which acts on orbital states in the same way spin operator $\vec{s}$ on spin states. The Hilbert space on each site consists of 4 basis states, which we choose as $\mid s_z, \tau_z\rangle$. We label them as

$$\begin{aligned} \mid 1\rangle =& \mid \frac{1}{2}, \frac{1}{2}\rangle, \mid 2\rangle = \mid -\frac{1}{2}, \frac{1}{2}\rangle, \\ \mid 3\rangle =& \mid \frac{1}{2}, -\frac{1}{2}\rangle, \mid 4\rangle = \mid -\frac{1}{2}, -\frac{1}{2}\rangle. \end{aligned} \tag{1}$$

These basis states form a fundamental representation of $SU(4)$. The conventional $SU(4)$ generators S^n_m acts on the basis state $\mid l\rangle$ according to $S^n_m \mid l\rangle = \delta_{n,l} \mid m\rangle$, and satisfies $\sum_m S^m_m = 1$ and $(S^n_m)^\dagger = S^m_n$. The Lie algebra is given by

$$[S^n_m, S^l_k] = \delta_{n,k} S^l_m - \delta_{m,l} S^n_k. \tag{2}$$

s_α, τ_β, and the product $s_\alpha \tau_\beta$ can all be expressed as linear combinations of the S^n_m [7]. For instance, $2s_z = S^1_1 - S^2_2 + S^3_3 - S^4_4$, $s^\dagger = S^2_1 + S^4_3$, and similarly for the orbital operators. Ignoring anisotropy and Hund's rule, the Hubbard model with double orbital degeneracy in the large U limit with 1 electron per site (1/4 filling) gives rise to the effective Hamiltonian

$$H = \sum_{i,j} J_{ij} S^n_m(i) S^m_n(j), \tag{3}$$

with repeated indices n, m summed. H is clearly invariant under global $SU(4)$ transformation. We have argued [7] that the ground state of H on many 2D lattices should be a flavor liquid. It is however useful to study the possible excitations assuming the ground state has spontaneous symmetry breaking. This is because i) while the ground state for the fundamental representation is disordered, it may have LRO for higher representations ii) such a study can provide information about the stability of certain classical ground states against quantum fluctuations; and iii) LRO can exist in $3D$ and/or away from the $SU(4)$ limit; and this analysis will allow insight as to what aspects of those excitations are due to proximity to $SU(4)$ symmetry.

Just as in the $SU(2)$ case, it is useful to generalize the $s = 1/2$ problem to general s, we can also consider representations other than the fundamental representation for $SU(4)$. In particular, we generalize it to representations denoted by Young tableaux with a single row but arbitrary columns M. In the limit $M \to \infty$, the non-commutativity between the S^n_m can be ignored, and the operators become c-numbers. For general M, the Lie algebra (2) can be exactly reproduced by a generalized HPT using 3-bosons b^m_n at each site, with $n \neq m$. The vacuum of these bosons is the state with $S^m_m = M$, where the choice of m is formally arbitrary, but in practice is taken to be the ordering "direction" of the classical ground state. The generalized HPT is defined as (for $n, l \neq m$):

$$\begin{aligned} S^m_m &= M - \sum_{n\neq m} b^{m\dagger}_n b^m_n, \\ S^m_n &= b^{m\dagger}_n \sqrt{M - \sum_{l\neq m} b^{m\dagger}_l b^m_l}, \\ S^l_n &= b^{m\dagger}_n b^m_l. \end{aligned} \tag{4}$$

Eq. (4) enables us to carry out the linear flavor wave theory by expanding the Hamiltonian in power of 1/M to quadratic order in b and/or $b^\dagger$ and then set $M = 1$ for the present physical system.

First consider the "ferromagnetic" (FM) case, $J_{ij} \leq 0$ in (3). The classical ground state, which is also the exact ground state, is given by $S^n_n(i) = M\delta_{n,1}$. Expansion of (3) leads to

the flavor wave Hamiltonian (omitting the superscript 1),

$$H_{fw}^{FM} = \sum_{\langle ij\rangle,l} |J_{ij}| \left([b_l^\dagger(i) b_l(i) - b_l^\dagger(i) b_l(j)] + i \leftrightarrow j \right),$$

where $l = 2, 3, 4$. This is comprised of three independent boson Hamiltonian corresponding to "FM" spin, orbital, and spin-orbital waves.

For the AF $SU(4)$ system, the classical ground state may be obtained by replacing the operators in (3) by their expectation values with respect to states of the form $|\Psi\rangle = \prod_i |\phi_i\rangle_i$, so that $\left\langle S_l^k(r) \right\rangle = \left\langle \phi_r \left| S_l^k \right| \phi_r \right\rangle$; and then minimizing the energy. Since $\sum_{nm} \langle S_n^m(i)\rangle \langle S_m^n(j)\rangle \geq 0$, the classical minimum for each bond is obtained by having the two sites connected satisfy $\langle \phi_i | \phi_j \rangle = 0$. This can be achieved by having any two sites connected by a bond having different flavors. Assuming this can be done for all bonds (unfrustrated), the classical ground state is then identical to that of the AF 4-state Pott's model. For concreteness, let us consider for now the square lattice with n.n. coupling J and next n.n. coupling J', with $J > 2J'$. Out of the degenerate manifold of classical ground states, we select for illustration the one obtained by dividing the lattice into 4 interpenetrating square sublattices and having all sites on sublattice α be in the flavor state $|\alpha\rangle$ (see Fig. 1a), i.e.$\langle S_n^m(i)\rangle = \delta_{nm}\delta_{n\alpha_i}$, where α_i denotes the sublattice the site i is on. Expanding (3) to the leading order in M, we have $H = \sum_{mn} H_{mn}$, where mn are the pairs of the 4-states in (1) with $m \neq n$, and

$$H_{mn} = \sum_{i,j} J_{ij} \{ b_n^{m\dagger}(i) b_n^m(i) + b_m^{n\dagger}(j) b_m^n(j) + [b_n^{m\dagger}(i) b_m^{n\dagger}(j) + h.c.] \}, \tag{5}$$

where i and j are summed over all the sites in sublattices m and n respectively. Note that the different pairs of H_{mn} are decoupled as a consequence of the $SU(4)$ symmetry. A boson b_n^m at sublattice m only coupled to its "mirror" boson b_m^n at sublattice n. This allows us to solve each H_{mn} separately, and H_{mn} is a simple Bogoliubov problem identical to the usual spin wave theory in the HAH. The spin (s), orbital (τ), and spin-orbital ($s\tau$) wave dispersions are given by $\omega_s(\vec{k}) = 2J\sqrt{1-\cos^2 k_y}$, $\omega_\tau(\vec{k}) = 2J\sqrt{1-\cos^2 k_x}$, and $\omega_{s\tau}(\vec{k}) = 4J'\sqrt{1-\cos^2 k_x \cos^2 k_y}$. Note that the spin and the orbital excitations are 1D in spite of the

underlying translationally invariant 2D ordering pattern. The 1D density of states can be understood from the ordering pattern of the classical ground state in Fig. 1a. There is no energy cost if we take any vertical or horizontal line and interchange the flavors on the two sublattices (creating a line defect). The disordering effect due to quantum fluctuations of these 1D like excitations should act just like those in 1D quantum spin chains and destroy the LRO. Of course, a linearlized theory can only be used as a guide to the true situation. [12,13] Nevertheless the result here is consistent with the previous results that the ground state of the AF $SU(4)$ model is a flavor liquid on many 2D lattices. In the limit $J' \to 0$, point defects (e.g., flip flavor from 1 to 4 on a site) can be created with no cost in energy, thus $\omega_{s\tau} \to 0$. Note that for $J' > 0$, the classical ground state does not have finite entropy per site in the thermodynamic limit, but the classical order is destroyed by inifnitesimal quantum fluctuations (all $M < \infty$) .

While the details of the above depend on the particular ordered state we choose, the central results of no mixing between different waves and existence of the excitations with reduced dimensionality are consequences of the $SU(4)$ symmetry. We now study the effects of deviation from this symmetry. We will illustrate the physics using an example which is of particular interest, the 2D honeycomb lattice, corresponding to a single plane of V_2O_3, whose magnetic behavior Castellani et. al. [5] sought to explain by invoking orbital degeneracy. The experimentally determined magnetic structure of the ordered state is consistent with that of 4-sublattice ordering discussed above with the lattice appropriately modified [3], as shown in Fig. 1b. The full Hamiltonian when Hund's rule and anisotropy are taken into account is quite complicated. Since we are principally interested in how the physics changes away from the $SU(4)$ symmetry in ways that are independent of the precise model, we will ignore Hund's rule and consider a Hamiltonian including orbital anisotropy,

$$H^{hc} = \frac{2}{U} \sum_{\langle ij \rangle} S_m^n(i)\, S_{n'}^{m'}(j) t_{nn'}^{(ij)} t_{mm'}^{(ij)}, \tag{6}$$

where $\langle ij \rangle$ denotes n.n. pairs, m, n, m', n' are summed from 1 to 4, and $t_{mn}^{(ij)}$ is the hopping integral from state $\mid m\rangle$ at site i to state $\mid n\rangle$ at site j [11]. Because of orbital anisotropy,

$t_{mn}^{(ij)}$ is diagonal in spin space, but not in orbital space. The hopping matrix for different orientational bonds $\langle i'j'\rangle$ and $\langle ij\rangle$ are related [5] by a rotational transformation. Note that while $t_{mn}^{(ij)}$ can be made diagonal on any one bond, it cannot be simulataneously done for all bonds. We choose the atomic orbitals with the hopping matrix diagonal for the "horizontal" bonds with eigenvalues $t_<$ and $t_>$. The quantity $\eta = \frac{t_> - t_<}{t_> + t_<}$ is a measure of the strength of the anisotropy. Expansion of the Hamiltonian in M leads to the flavor wave Hamiltonian,

$$H_{fw}^{hc} = \frac{2}{U} \sum_{\langle ij\rangle} \sum_{\alpha \neq \alpha_i} [b_\alpha^{\alpha_i \dagger}(i) b_\alpha^{\alpha_i}(i) t_{\alpha_i \alpha_j}^{(ij)} t_{\alpha \alpha_j}^{(ij)} + \sum_{\beta \neq \alpha_j} (b_\alpha^{\alpha_i \dagger}(i) b_\beta^{\alpha_j \dagger}(j) t_{\alpha_i \beta}^{(ij)} t_{\alpha_j \alpha}^{(ij)} + h.c.)]. \tag{7}$$

Consider first the $SU(4)$ limit, $t_{mn}^{(ij)} = t\delta_{m,n}$, namely $\eta = 0$. H_{fw}^{hc} is reduced to the form of (5) with $J = 2t^2/U$, and the spin, orbital, and spin-orbital waves are decoupled. The different connectivity of the honeycomb lattice and the ordering pattern in Fig. 1b results in some modifications from that in the sqaure lattice in the dispersion of the excitations. Flavors 1 and 4 (or 2 and 3) are connected as zig-zagging vertical chains, and spin-orbital waves are 1D like with dispersion $\omega_{s\tau}(\vec{k}) = J \sin(\sqrt{3}|k_y|/2)$. Flavors 1 and 2 (or 3 and 4) are unconnected, and spin waves are localized on a site and have zero excitation energy, $\omega_s = 0$. Flavors 2 and 4 (or 1 and 3) are connected as two-site pairs which are decoupled from other pairs. The orbital modes (b_1^3, b_2^4) are thus also localized with $\omega_\tau = 0$.

With anisotropy, the situation changes significantly. Spin is still conserved, but orbital no longer. Thus, spin modes (b_2^1, b_4^3) and spin-orbital modes (b_4^1, b_2^3) are now mixed, and therefore both neutron scattering active. However, for convenience, we continue to label them as spin and spin-orbital modes. Orbital information can thus be obtained indirectly from neutron scattering experiment. For H^{hc}, the dispersion of the spin-orbital mode remains 1D like. The lack of coupling between zig-zagging vertical chains can be understood simply as follows. Because the hopping matrix is diagonal on the horizontal bonds, the coupling between the two sites connected by such a bond in (7) contains only terms of the form $S_m^n(i)\, S_n^m(j)$. Thus, this coupling conserves $S_n^n(i) + S_n^n(j)$, and hence the chains are decoupled for spin-orbital and also spin modes for the same reason as the $SU(4)$ case. For spin wave mode, the dispersion in fact remains dispersionless with zero excitation energy, indicating

they remain localized, albeit no longer on a single site. Pure orbital excitations remain localized on the two sites of each horizontal bond and decoupled from others.

In Fig. 2a we show the energy dispersion as a function of k_y for the spin and spin-orbital modes, which are the modes that can be probed by neutron scattering. Excitation energy is shown in units of $J = 2t_>^2/U$. There is no dispersion with k_x since the chains are decoupled for these modes. The four purely spin modes $(b_2^1, b_1^2, b_4^3, b_3^4)$ in the $SU(4)$ limit remains dispersionless with anisotropy but with some spin-orbital characteristics mixed in. The four spin-orbital modes$(b_4^1, b_3^2, b_2^3, b_1^4)$, which for $SU(4)$ were all degenerate, are split with anisotropy into two two-fold degenerate branches with the upper branch no longer a Goldstone mode as $k_y \to 0$. If the effects of Hund's rule are included with effective strength J_H, chains will be coupled, and all the modes will have 2D dispersion. Furthermore, any accidental degeneracy and/or zero energy excitations will be removed. The spectrum will consist of two degenerate Goldstone spin waves whose dispersion is linear for small k with a slope whose scale is determined by J_H; two other degenerate spin waves with a gap and dispersion also detemined by J_H; two degenerate spin-orbital waves with a gap and dispersion in k_x of order J_H, but whose dispersion in k_y is of order J; two other degenerate spin-orbital modes with gap of order $J\eta$, and k_x and k_y dispersion of order J_H and J respectively. The twofold degeneracy in each case is required by the remaining $SU(2)$ spin symmetry of the Hamitonian. These features are qualitatively in agreement with inelastic neutron scattering data on V_2O_3 which shows that the effective in-plane spin-spin coupling between parallel spins to be considerably weaker than that between antiparallel ones. Details of the calculation including Hund's rule and interplane coupling of V_2O_3 will be discussed in a later publication.

Because of the mixing between spin and spin-orbital waves, neutron scattering experiments will couple to all the modes mentioned above. The intensity of inelastic neutron scattering cross section is proportional to $Im\,\chi(\vec{q},\omega)$, where χ is the transeverse susceptibility. Within the flavor wave approximation, the spin lowering operators $S^-(i) = S_2^1(i) + S_4^3(i)$, which is $b_{\alpha_i+1}^{\alpha_i\dagger}$ if $i \in$ sublattice 1 or 3, and $b_{\alpha_i-1}^{\alpha_i}$ if $i \in 2$ or 4, and similarly for the raising

operator. The relative intensity can thus be calculated straightforwardly using the Bogoliubov transformation. We show an example of this for $T = 0$ in Fig. 2b for $\eta = 0.43$ and without Hund's rule. For finite T, the intensity of each branch needs to be multiplied by $(1 + 2/(e^{\beta\omega} - 1))$.

In summary, the Holstein Primakoff transformation has been generalized to develop a quantum flavor wave theory for spin systems with orbital degeneracy. In addition to spin and orbital bosonic excitations, proximity to $SU(4)$ symmetry gives raie to a third mode of boson excitations observable by neutron scattering.

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FIGURES

FIG. 1. Four-sublattice classical ground states considered in the text. a) Square lattice; b) Honeycomb lattice.

FIG. 2. a) Spin (triangules) and spin-orbital (circles and squares) wave spectra with orbital anisotropy $\eta = 0.43$ for the honeycomb lattice as a function of k for wavevector (k_x, k). Dashed curve shows the (4-fold degenerate) spin-orbital excitations in the $SU(4)$ limit. b) Calculated neutron scattering intensities (arbitray units) for the spin (triangles) and spin-orbital (circles and squares) modes

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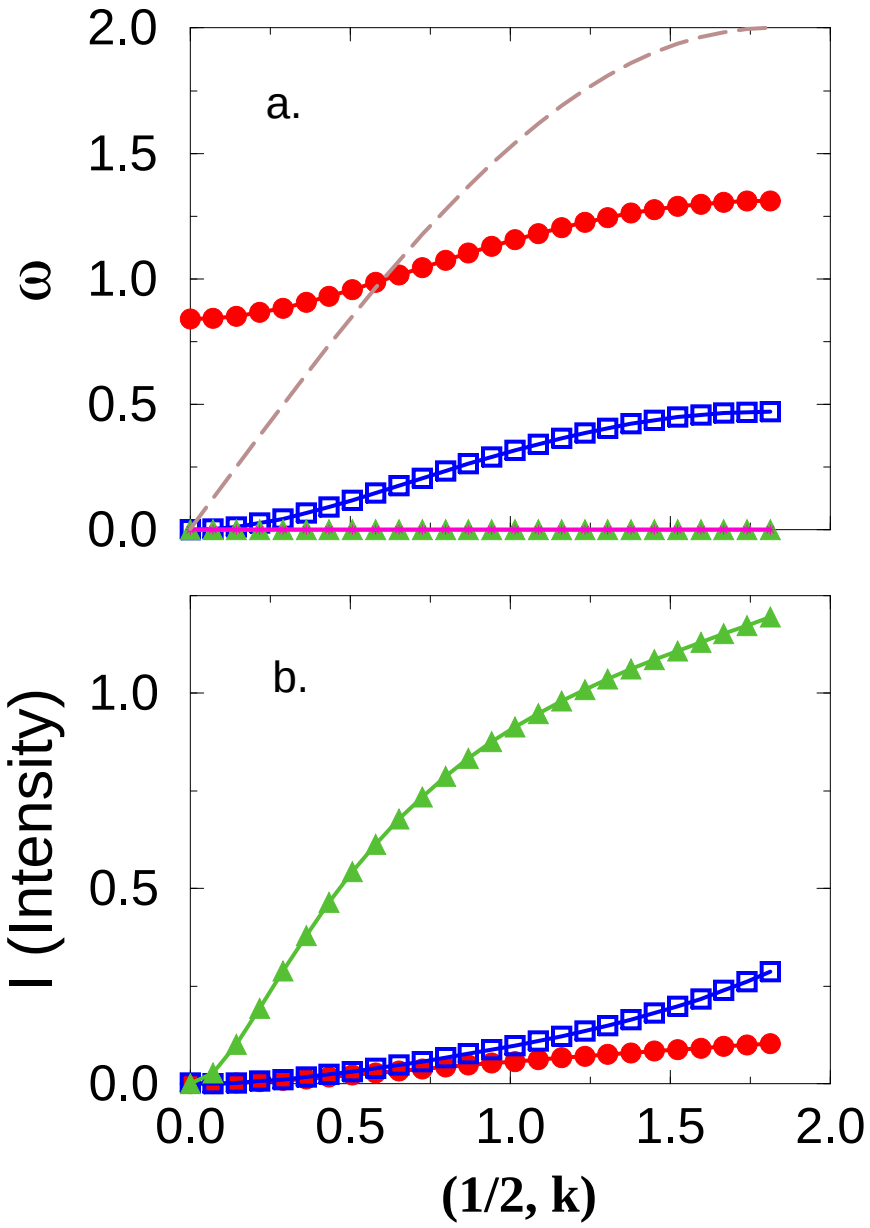
a.
b.
ω
I (Intensity)
(1/2, k)
0.0
0.5
1.0
1.5
2.0